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As part of the Global Nuclear Energy Partnership (GNEP) and the Advanced Fuel Cycle Initiative (AFCI), the US Department of Energy (DOE) is participating in an international collaboration to irradiate prototypic actinide-bearing transmutation fuels in the French Phenix fast reactor (FUTURIX-FTA experiment). The INL has contributed to this experiment by fabricating and characterizing two compositions of metallic fuel; a non-fertile 48Pu-12Am-40Zr fuel and a low-fertile 35U-29Pu-4Am-2Np-30Zr fuel for insertion into the reactor. This paper highlights results of the microstructural analysis of these cast fuels, which were reasonably homogeneous in nature, but had several distinct phase constituents. Spatial variations in composition appeared to be more pronounced in the low-fertile fuel when compared to the non-fertile fuel.

I. INTRODUCTION

The GNEP program is an international collaboration to develop comprehensive and proliferation resistant nuclear fuel cycle technologies. As part of this effort, viable fuel forms of minor actinide bearing fuels need to be developed so that long-lived fission products from recycled spent nuclear fuel (SNF) can be eliminated in a "burner" type reactor. Development of such technologies has the potential to greatly reduce the volume of high-level radioactive waste that will need to be disposed of in a permanent repository.

Metallic fuels for were the first fuels developed for fast reactors and have several performance advantages over their ceramic counterparts [1]. Metallic fuels have exceptional transient behavior, excellent thermal conductivity and a more straight forward reprocessing path which does not separate out pure plutonium from the process stream[2]. Because of their low melting point, binary U-Pu alloys cannot be used as a fuel in a sodium metal cooled fast reactor. A variety of alloying elements were experimented with to increase the melting point, and Zr was chosen primarily because it has a low neutron capture cross section and also serves to suppress interdiffusion between the fuel constituents and the cladding [3].

In the recycle process, the SNF is separated into U, fission products and a process stream containing ~90% Pu and 10% minor actinides (Am, Np, Cm)[4]. It is these constituents that will make up the transmutation fuels. The properties of this fuel form will most likely be significantly different and potentially more complex than traditional U and U-Pu based fuel forms. In addition, the burn-up levels desired (20 at-%) in order to minimize the process losses associated with multiple recycle passes, will further challenge the fuel performance. In order to establish both technical and safety cases for these fuel forms, extensive studies on thermophysical properties and irradiation performance are being conducted under the AFCI program. This paper presents results on the microstructural characterization of as cast metallic fuels that are being inserted into the French fast reactor Phenix in order to evaluate extended burn-up fuel performance.

II. EXPERIMENT

Arc casting was conducted in an inert gas (argon) glovebox using an electric discharge plasma arc melter with a suction extraction casting method. The feedstock materials were melted and thoroughly homogenized. A ZrO_2 coated quartz tube was then dipped into the molten liquid and the liquid was drawn up into the tube using a syringe. Images of the cast fuel slugs are shown in Figure 1. The slug dimensions for the fuels cast in this study were approximately 5 cm in length and 4.9 mm in diameter. Visual inspection of the slugs was made following cooling and removal of the samples from the quartz tubes. Slugs that were visually void free were transferred to a second air glovebox for metallographic preparation.

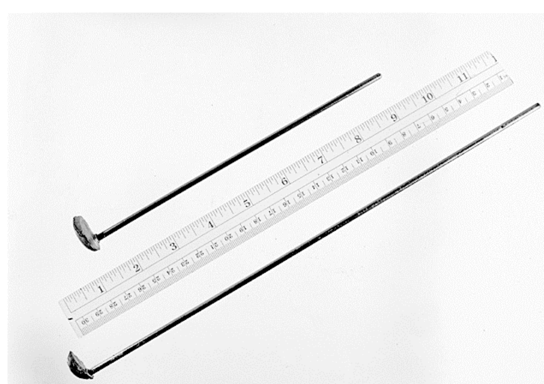


Figure 1 image of as cast fuels slugs

Table 1 Nominal composition of fuel slug castings

Fuel	U	Tolerance	Pu	Tolerance	Am^{241}	Tolerance	Np^{237}	Tolerance	Zr
DOE 1	35	±5%	29	±5%	4.0	±10%	2.0	±10%	30
DOE 2	0.0	0.5%	48	±5%	12.0	±10%	0.0	1.5%	40

Table 2 Isotopic content of plutonium in cast alloys

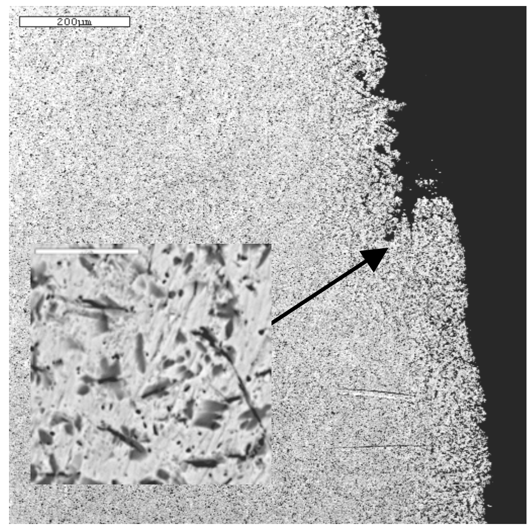
Pu^{238}	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}
0.05	82.54	16.5	0.57	0.34

Four samples were analyzed in this study. Three of the samples had a nominal composition of 48Pu-12Am-40Zr but were taken from different casting heats, and the fourth sample had a nominal composition of 35U-29Pu-4Am-2Np-30Zr. Table 1 contains the nominal compositions and allowable chemistry limits of the elements in the fuels, while Table 2 contains the isotopic concentrations of the plutonium used for the fuels. The SEM sample surface was coated with a thin layer of palladium to eliminate charging in the SEM before being transferred into the microscope. Sample analysis was conducted in a Zeiss 960a SEM equipped with both Energy Dispersive and Wavelength X-Ray detectors. The microscope was operated at an accelerating voltage of 30kV. Because no standards existed in the spectrometer software for transuranic elements, only relative concentrations of these elements could be determined..

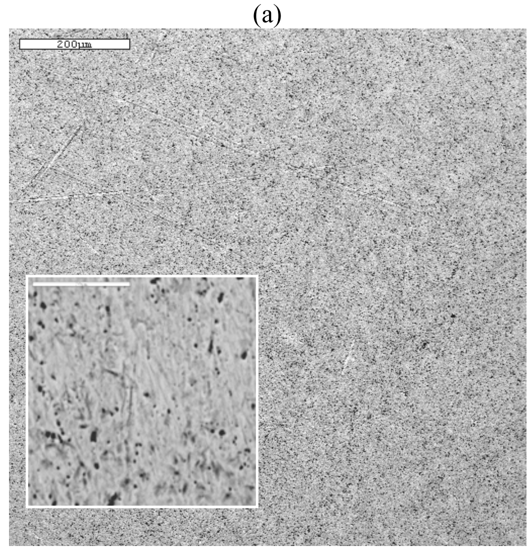
III. RESULTS AND DISCUSSION

III. A. 48Pu-12Am-40Zr Alloy (Non-Fertile)

For the non-fertile fuels, all three fuel slug heats that were nominally the same composition had similar microstructure. Figure 2 is an SEM image showing two areas of a fuel slug cross-section, one on the perimeter and one in the center of the slug. A region approximately 50 – 150 microns in width around the perimeter of the sample had a microstructure evidently different from the interior of the fuel as illustrated in the insets of the figure. Large blocky precipitates found in the perimeter region are not observed in the fuel interior. The origin of the differing microstructure is thought to come form ingress of oxygen and other impurities from either a ZrO_2 coating on the quartz casting mold or the mold itself. The rest of the fuel interior is relatively homogenous containing two phases; a secondary phase of small precipitates enriched in Zr and depleted in actinides surrounded by a matrix phase containing a high density of fine pores (revealed in the high magnification image shown in the inset).



(a)



(b)

Figure 2 Backscatter electron SEM images of the non-fertile 48pu-12Am-40Zr fuel sample: a) perimeter of fuel slug b) center of slug. Inset provides higher magnification view showing finer characteristics of the microstructure.

X-ray diffraction data obtained prior to the SEM characterization showed only the matrix phase, which was identified as having a cubic delta Pu structure (Fm-3m;a=4.57). The absence of additional phases in the X-ray data suggests the size and volume fraction of the Zr rich phases in the alloy are not substantial enough to show up in the diffraction pattern.

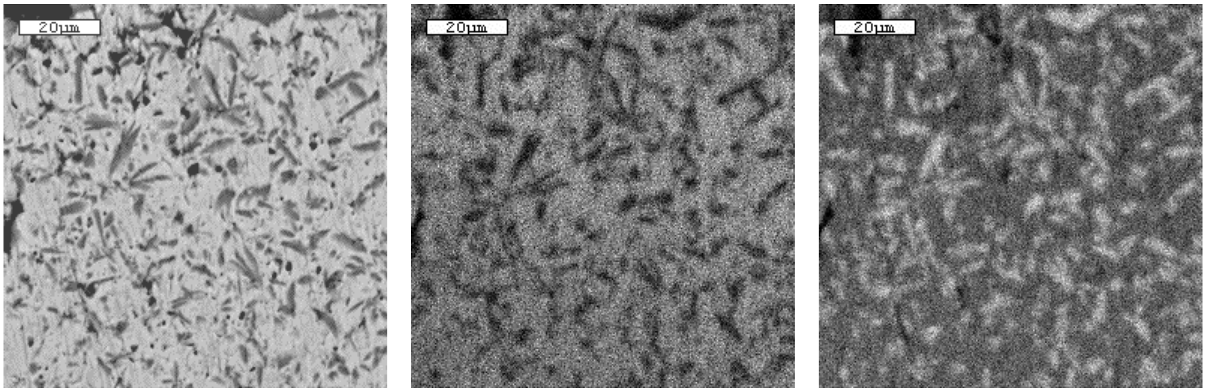


Figure 3 X-ray maps of the microstructure in the non-fertile 48Pu-12Am-40Zr fuel perimeter showing phases enriched in Zr and depleted of Pu. Bright contrast features in the maps indicate a relative enrichment of that element compared to the surrounding area, while dark contrast features represent the fine porosity in the fuel.

SEM elemental X-ray maps shown in Figure 3 reveal the blocky precipitate in the outer rim are enriched in Zr. These precipitates are also depleted in actinides relative to the matrix. The X-ray maps also revealed the presence of a Si-rich rind on the exterior perimeter of the sample and Si-rich precipitates throughout (Figure 4) . This is most likely due to the fact that the alloys are frequently cast, remelted and cast again thereby distributing the Si from the quartz tube throughout interior of the sample. The X-ray maps also revealed that, although having a different morphology than the perimeter phase, the secondary phase in the interior of the fuel is enriched in Zr and depleted of actinides relative to the matrix.

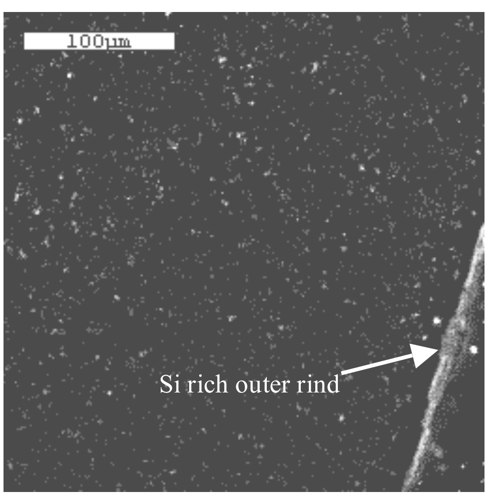


Figure 4 Si X-Ray Map of 48Pu-12Am-40Zr fuel sample.

III.B. 35U-29Pu-4Am-2Np-30Zr Alloy (Low-Fertile)

As with the non-fertile fuel composition, there is an outer rind on the surface of the sample and a region ~50 to 150 microns around the perimeter containing larger second phase precipitates. A lower magnification image showing the different phase regions is shown in Figure 5. Moving toward the center, the morphology of the precipitates in the outer perimeter zone is somewhat different than the non-fertile fuel being more stringer-like than blocky as indicated in the backscatter electron image of Figure 6. The microstructure adjacent to the perimeter zone appears homogenous and, as with the previous samples, contains two phases consisting of small precipitates surrounded by a matrix phase as well as a density of fine pores. In the central region of the slug, backscatter electron imaging, which emphasizes compositional differences, reveals a change from rather uniform composition in the matrix phase to minor compositional variations throughout the phase. The second phase precipitates appear to be concentrated in the matrix regions having a lower Zr content.

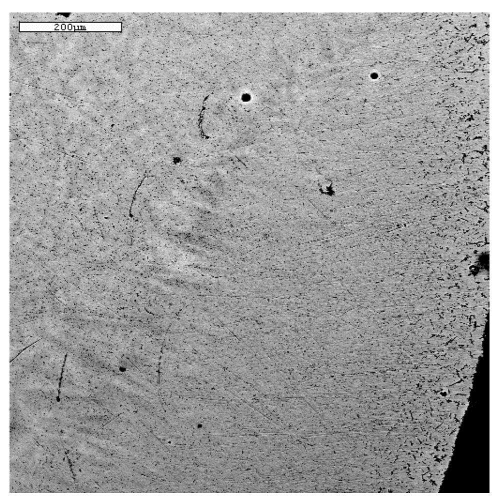
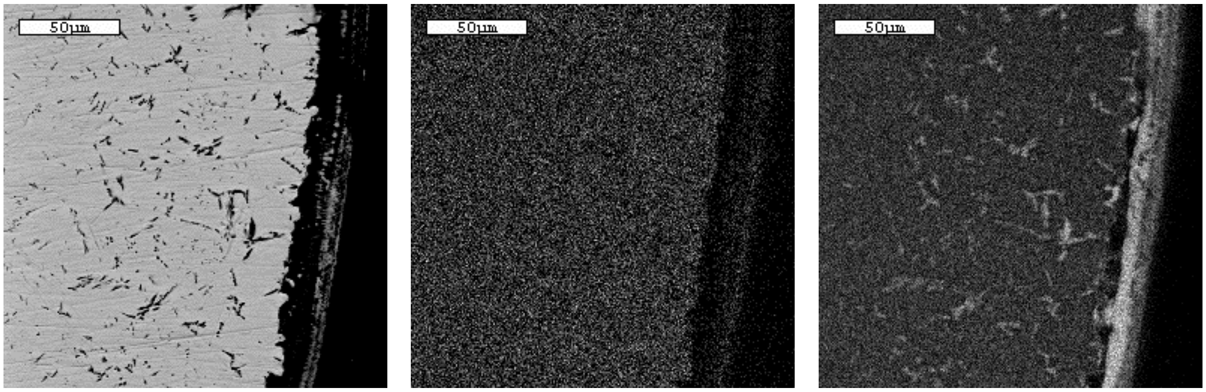
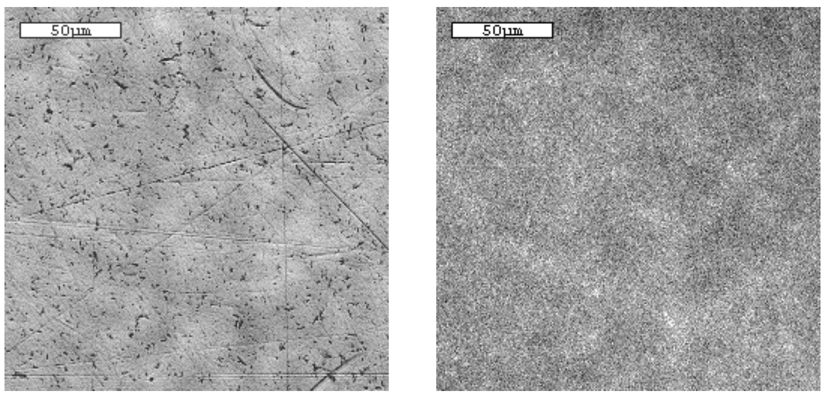


Figure 5 BSE image encompassing perimeter region and interior of 35U-29Pu4Am-2Np 30Zr.



BSE image Pu X-ray map Zr X-ray map

Figure 6 X-ray maps of the fertile 35U-29Pu4Am-2Np 30Zr fuel at the outer perimeter of the sample.



a) b)

Figure 7 a) Higher magnification BSE image of 35U-29Pu4Am-2Np 30Zr center region along with b) a Zr X-ray map showing minor variations in composition in the matrix phase.

X-ray diffraction shows the major phase constituent of the alloy to be hexagonal and modeled to the δ-UZr_2. Strong X-ray peaks from other phases were not present.

From X-ray mapping (Figure 6) of the perimeter zone, the second phase precipitates in this region are also enriched in Zr, however, depletion of actinides in this phase is not as evident as the non-fertile fuel. The outer surface surface rind is enriched in Zr and Si, indicating once again as with the non-fertile fuel, constituents from the quartz casting mold are adhering to the fuel. The higher magnification Zr X-ray map shown in Figure 7 reveals the contrast changes in the electron image are in fact due to spatial variations in the Zr concentration.

III. C. Non-Fertile vs Low-Fertile

In comparing the non-fertile and fertile fuels, neither one has significant local heterogeneities, such as large pores or cavities or inclusions that would indicate non-uniform distribution of elemental constituents. Although porosity was present in the castings of both alloys, the porosity was on a extremely fine scale. Both alloys also exhibit an outer perimeter zone with larger Zr-rich second phase precipitates and smaller Zr-rich precipitates in the fuel interior. Depletion of the actinide elements in the outer zone second phase precipitates appears less pronounced in the low-fertile fuels compared to the non-fertile fuels. The major microstructural difference between the two alloys is there appears to be more segregation of Zr in the matrix phase of the low-fertile alloy. More detailed studies to quantify the compositions of the separate phases will need to be performed in order to clarify these differences.

IV. CONCLUSIONS

Cast metallic fuels destined for the Futurix-FTA irradiation experiment were examined in this study. The following conclusions can be made from the study.

1. Both non-fertile and fertile phases contain a surface rind rich in Zr and Si, indicating adherence of

constituents from the ZrO2 coated quartz mold used for the casting the fuels.

2. Diffusion of mold constituents into the cast fuel promotes the formation of larger Zr-rich phases in a zone approximately 50 – 150 µm around the outside perimeter of the fuel.

3. The non-fertile fuel interior consists of two phases, small Zr-rich precipitates within a matrix containing a high density of fine pores.

4. The general characteristics of the low-fertile fuel is similar to the non-fertile fuel, having a Zr and Si-rich outer rind, and an outer permiter zone containing larger Zr-rich precipitates.

5. The interior of the low-fertile fuel appears to be composed of two regions with the principle difference being the matrix phase in the central region of the fuel has spatial variations in Zr-content compared to the region closer to the outside of the fuel.

V. ACKNOWLEDGMENTS

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VI. REFERENCES

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